Response to Referee #2

The authors thank the reviewer for the comments that improve the quality of the paper. The detailed responses are given as follows. The reviewer comments are shown in italic fonts, the responses are in regular font, and the revised text is in bold font.

In this paper, Li et al. have modified the CMAQ model to take into account the impacts of water partitioning and polarity of organic compounds on SOA formation. The model was applied over Eastern China to estimate the regional and seasonal impacts of these modifications on SOA and the aerosol water content. This study may have potential to contribute in the organic aerosol modeling field but major revisions needs to be done before publication. In particular, I have several concerns regarding the validity of the scientific methodology used and the presentation of the study. Therefore, I would recommend publication only if these comments will be addressed and fundamental changes will be contacted.

Major comments

Comment 1: *Page 4 line 94: The majority of current CTMs have replaced the 2-product model with the VBS approach. Please make this clear and refer to the 2-product model of Odum et al. (1996) for historical reasons.*

Response 1: The text has been revised in the manuscript (L88-95) to make this clear:

"**The formation of condensed organic products is commonly represented by lumped surrogate SVOCs in a 2-product model with volatilities and SVOC yields fitted to chamber experiments (Odum et al., 1996). To better represent the volatility of primary organic aerosol (POA) and the multi-generation oxidation of SVOCs to a wider range, Donahue et al. (2006) proposed the volatility basis set (VBS) model in which the mass yields of SVOCs are fitted to a fixed number of volatility bins (usually 0.01-105 μg m-3). The VBS model has been adopted by several CTMs (such as WRF-Chem, GEOS-Chem, etc.).**"

Comment 2: *Page 6, line 162: Please add the appropriate references to support the nonvolatile nature of the products by these oxidation pathways.*

Response 2: Formation of these non-volatile SOA was traditionally treated in the CMAQ model, except for dicarbonyls, IEPOX, and MAE that are assumed to form SOA by irreversible reactive uptake in our model, as an upper-limit estimation of SOA from these precursors. The text has been revised to make this clear (L162-173 in the manuscript):

"The SOA module mostly follows Pankow et al. (2015). Two types of SOA as traditionally treated in CMAQ were considered, "semi-volatile" (SV) portion that formed via equilibrium absorption-partitioning of SVOCs, and

"non-volatile" (NV) portion that includes the oligomers and SOA formed via direct oxidation of aromatics at low-NOx. SOA from dicarbonyls, IEPOX, and MAE were formed by irreversible reactive uptake and categorized as NV-SOA in the current model as well. Some studies investigated SOA from glyoxal, methylglyoxal, and IEPOX using detailed reactions and reversible pathways in models or observed as reversible processes in chamber experiments, leading to a relatively lower SOA yield compared to the surface-controlled irreversible uptake (Lim et al., 2013; Knote et al., 2014; Galloway et al., 2009; El-Sayed et al., 2018; Budisulistiorini et al., 2017). The non-volatile assumption used in this paper allows an upper-limit estimation of the importance of these additional SOA formation pathways."

Comment 3: *Page 6 line 149: There is no discussion in the methodology about the observations and the statistical analysis metrics used to evaluate the model performance. Especially for the OA observations, there is no reference provided or description of the methods used.*

Response 3: Thank you for pointing this out. We have included details of observation data, statistical analysis metrics for both meteorology and aerosols in the revised manuscript:

In L265-276

"**The meteorological inputs and emissions have been used in several previous publications. Model performance on meteorological parameters (temperature and RH), gaseous species and gas and aerosol concentrations have been extensively evaluated (Hu et al., 2016; Hu et al., 2017; Qiao et al., 2018; Shi et al., 2017). A summary of the model performance related to this study is provided below. Observed meteorological data were obtained from the National Climatic Data Center (ftp://ftp.ncdc.noaa.gov/pub/data/noaa). Observations of OC at two urban locations, Beijing (Cao et al., 2014; Wang et al., 2015) and Guangzhou (Lai et al., 2016) and OA in Beijing (Sun et al., 2014) during January of 2013 as well as surface $PM_{2.5}$ at several monitoring sites during July of 2013 from China National Environmental Monitoring Center (http://113.108.142.147:20035/emcpublish/) were used to evaluate model estimates of aerosols. Details of measurement methodology and uncertainties of observations are listed in the corresponding references.**"

In L278-282

"**Table 1 lists model statistics of mean observation (OBS), mean prediction (PRE), mean bias (MB), gross error (GE) and correlation coefficient (R) based on WRF and observations at monitoring sites located in 8 sub-regions of the domain (Figure S1) during January and July of 2013. The benchmarks for the MM5 model (another meteorology model) of 4-12km horizontal resolution suggested by Emery et al. (2001) are also listed in**

the table.”

In L294-296

“**Overall, the mean fractional bias (MFB) and mean fractional error (MFE) of OC are -0.20 and 0.27, within the criteria (MFB≤±0.6; MFE≤0.75) suggested by EPA (2007).**”

In L308-309:

“**The overall MFB and MFE of OA during January are -0.28 and 0.54, within the criteria (MFB≤±0.6; MFE≤0.75) suggested by EPA (2007).**”

Comment 4: *Page 7 line 179: More details are needed here. (1) How the model defines the low NOx and high NOx conditions? Which compounds each of the lump species represent? What is the difference between the lumped species of the same precursor (e.g., BNZ1, BNZ2, BNZ3)? (2) Can you include the aerosol yields for each lumped species in tables S1 and S2? Are these aerosol yields NOx-dependent? (3) What does the SVP stands for in Tables S1 and S2?*

Response 4: (1) SOA formation in CMAQv5.0.2 is based on the frame of a previous version 4.7.1. All the details about “high” and “low” NO_x conditions (based on chamber experiments of corresponding VOCs), lumping species and method of each precursor, and the yields of precursors from parent VOCs have been documented by Carlton et al. (2010) and summarized in the revised supplemental materials as following:

“**The CMAQ model treats high and low NOx SOA formation pathways during OH oxidation by allowing the lumped RO2 radical to competitively react with HO2 and NO. Using the lumped ARO1 species as an example, an SOA formation specific RO2 radical ARO1RO2 is added as a gas phase reaction product with OH:**

$$\mathbf{ARO1 + OH \rightarrow ARO1RO2 + products}$$

The ARO1RO2 can react with both HO2 and NO, as shown in the following two reactions:

$$\mathbf{ARO1RO2 + HO2 \rightarrow HO2 + TOLNRXN; \quad k1}$$

$$\mathbf{ARO1RO2 + NO \rightarrow NO + TOLHRXN; \quad k2}$$

Details of the determination of the rate constants can be found in Carlton et a. (2010). The TOLNRXN and TOLHRXN are counter species that track how much ARO1 is reacted through low NOx and high NOx pathways, respectively, in one gas chemistry time step. The concentrations of these counter species are passed into the aerosol module to calculate the formation semi-volatile products (TOL1 and TOL2) in the high NOx pathway and non-volatile products (TOL3) in the low NOx pathway, using the mass-specific yields, as listed in Table S1 and S2. Equilibrium partitioning of TOL1 and TOL2 in the gas phase and their counterparts ATOL1 and ATOL2 in the organic phase are affected by temperature and the amount of absorbing organics in the aerosol phase. Similar

treatments are applied to the other lumped aromatic compounds ARO2, with xylene as a representative and most abundant species in that group, and to benzene. SOA formation from lumped long-chain alkene species ALK5, and isoprene and monoterpenes are not considered as NO_x dependent and are represented by equilibrium partitioning of one or two semi-volatile oxidation products. Details of the mass-specific yields of semi-volatile products and other related parameters can be found in Table S1 and S2."

We revised the text in (L188-190) to make it clear:
"**More details about the lumped precursors such as formation conditions ("high" and "low" NOx), lumping species and method, and yields from parent VOCs can be found in Carlton et al. (2010) and summarized in SI.**"

(2) In the CMAQ model, the amount of SOA can form after a precursor reacts with OH, O_3 or NO_3 depends on the volatility of the products, which is temperature dependent, and the amount of the absorbing organics. The mass yields of the semi-volatile or non-volatile products are included in Table S1 and S2 in the revised supplementary materials. For more details, we refer the readers to Carlton et al. (2010) and the references therein.

(3) SVP is the saturation vapor pressure. We have explained this in the corresponding tables.

Comment 5: *Page 8, lines 194-198: (1) Does the absorbing phase of equation 1 includes only the water associated with the organics (from eq. 3) or it includes the total water (including the water associated with the inorganic aerosol components)? (2) Under high RH (higher than the organic/inorganic phase separation RH, SRH), the aerosol organic phase is well mixed with the inorganic salts and, therefore, the aerosol water associated with the inorganic constituents can also contribute to the SOA absorbing medium (Pye et al., 2017). Please clarify what you have assumed here and add the relative discussion.*
Response 5: (1) The absorbing phase of equation 1 only includes organic and water associated with organics when considering water-organic interactions. We explained this in the original text L195-196:
"In addition to organic compounds, water partitioning into OPM is enabled according to Eq 1 and Eq 2. In such a case, the absorbing phase in Eq 1 includes both organic aerosols and water partitioning into OPM."

(2) We didn't consider the mixing of organic and inorganic phase in this study and assumed that they are always two distinct aerosol phases without direct interactions. The phase separation RH (SRH) depends on the OM/OC ratio of the organic phase. Unlike the conditions modeled by Pye et al. (2017) for the

southeast US where SOA is often the dominant OA component, the winter episode we modeled is dominated by primary emitted organic aerosols thus with a relatively low OM/OC (~1.4-1.6). The SRH based on equation (7) of Pye et al. (2017) is ~97%-99%. The summer episode has more contributions of SOA to OA, with OM/OC~1.8, which corresponds to an SRH of 87%. Thus, we don't expect interactions of organic and inorganic phases to occur in high frequency to greatly influence the model results. We assumed no interactions between inorganic and organic phases in the current model. We have also revised text in L207-208:

"**In the current model, we assumed no interactions between the inorganic and organic phases.**"

Comment 6: *Page 8, Equation 3: This equation gives the volume of water associated with the organic fraction of the aerosol. However, ALW on the left-hand side refers to the mass of water. Please correct.*

Response 6: The equation has been corrected in the revised text L212-214:

"

$$\boldsymbol{ALW_{org} = \rho_w V_{org} \kappa_{org} \frac{a_w}{1 - a_w}} \qquad \textbf{(Eq3)}$$

where ρ_w is the density of water (assumed to be 1 g cm^{-1}), V_{org} is the volume concentration of organics, and a_w is the water activity (assumed to be the same as RH)."

Comment 7: *Page 8, lines 203-204: How do you estimate the hygroscopicity?*

Response 7: We used Eq3 to estimate the hygroscopicity. This has been clarified in the revised text in L214-216:

"**Since ALW_{org} in this study is calculated mechanistically using the partitioning theory, κ_{org} can be estimated by rearranging Eq3:**

$$\boldsymbol{\kappa_{org} = \frac{ALW_{org}}{\rho_w V_{org}} \times \frac{1 - a_w}{a_w}} \qquad \textbf{(Eq4)}$$

"

Comment 8: *Page 8, lines 204-205: How do you calculate the ALW? Are you using κ_{org} and the eq3? Do you use the kappa hygroscopicity to calculate the ALW? If so, how you estimate the kappa?*

Response 8: In this study, the ALW_{org} is independently calculated by mechanistically allowing water molecules to partition into the organic phase with UNIFAC calculated activity. In such a case, Eq 3 can be used to provide an independent estimation of κ_{org}. Linear regression analysis can be performed using the calculated κ_{org} against the model calculated O/C ratios, as shown in Figure 4. We have removed this sentence to avoid confusion.

Comment 9: *Page 9, lines 212-216: Add a reference to tables S1 and S2. Furthermore, why the values of OM:OC in tables S1/S2 are different than the values provided by your reference (Pye et al., 2017)?*
Response 9: We used OM:OC ratio in Pankow et al. (2015). There were mistakes in the original Table S1 and S2 and have been corrected in the revised supplemental materials. The text has also been revised in L228-229 as following:
"**The OM:OC ratio of each SOA component follows Pankow et al. (2015) as shown in Table S1-S2.**"

Comment 10: *Page 9 lines 217-218: Can you add the total size of the model domain?*
Response 10: The text has been revised in L231-233 as follows:
"**The simulation domain has a horizontal resolution 36 km × 36 km (100 × 100 grids) and a vertical structure of 18 layers up to 21 km, which covers eastern China as shown in Figure S1.**"

Comment 11: *Page 9 lines 233: (1) What boundary conditions are used? Please make a comment on how these can affect the simulation results. (2) I would recommend adding spatial maps of primary organic aerosol emissions and SOA precursor emissions and summarizing in a Table the domain average emission rates of POA and each SOA precursors.*
Response 11: (1) We used a predefined boundary profile in CMAQ that represents a clean continental condition. The northern and western boundaries, as well as areas to the further north and west, are mostly remote areas with much lower emissions. The mountains in the north and west part of the domain also limit the influence of emissions enter from the boundaries to the central part of the domain. The influence of marine air from the south and east boundaries is also small, as local emissions dominate the concentrations.

(2) A figure and a table showing emissions of POA and SOA precursors were added to the revised supplemental materials as Figure S2 and Table S4.

Comment 12: *Pages 9-10 lines 234-238: (1) This paragraph needs to be expanded and written in a separate section. In this section the authors should describe in more detail the following: i) Basecase simulation. Please explain how the default CMAQ is simulating POA and SOA and how different is this modelling configuration with the one the authors are testing, ii) Sensitivity simulations. Please explain in much more detail the sensitivity simulations conducted in this work. (2) In addition the authors say that they have conducted three sensitivity scenarios named S1, S2 and S3. In their manuscript they only show results from S3 and they never discuss the results of S1 and S2.*
Response 12: (1) Details of how default CMAQ simulates SOA formation through equilibrium partitioning of lumped semi-volatile products into the

organic phase (which include both POA and SOA) have been described by Carlton et al. (2010) and Hu et al. (2017) so we don’t think it is necessary to repeat it here. We expanded each simulation scenario in details in the revised text in L249-259:

“**Four scenarios are investigated in this study. The base case (BS) applies the default secondary organic aerosol module of CMAQ v5.0.1. In this case, no water partitioning into OPM is considered. Lumped semi-volatile products from the oxidation of various precursors partition into a single organic phase, which is considered as an ideal mixture of POA and SOA with γ_{org}=1. The water case (S1) includes water partitioning into OPM, which is again considered as an ideal solution (γ_{org}=1 and γ_{H2O}=1). The UNIFAC case (S2) considers the interaction between organic constituents with UNIFAC calculated activity coefficients ($\gamma_{org}\neq1$) but does not allow water partitioning into OPM. The combined case (S3) allows both water partitioning and interactions between all constituents (including water and organics) using UNIFAC calculated activity coefficients ($\gamma_{org}\neq1$ and $\gamma_{H2O}\neq1$).** ”

(2) The impacts of S1 and S2 were discussed in Section 4 of the original manuscript Page 16 L405-419. We also included a description of the outlines of the results and discussions in the original manuscript Page 6 L140-148. The text has been revised in L259-262 to make it clear:
“**The results of BS and S3 are used to examine the overall impacts of water partitioning into OPM and polarity of organics on SOA and ALWorg, as shown in Section 3.1-3.4. The separate influences of those two processes on SOA from S1 and S2 are discussed in Section 3.5.**”

Comment 13: *Page 10 line 240: The section “Model evaluation” is extremely problematic and raises questions on the validity of the modelling results given that the model evaluation is insufficient. More specifically: 1) Given that the CMAQ default configuration has been modified to consider the importance of water and organic compound polarity on SOA formation, an accurate evaluation of the updated model performance is needed. 2) You should compare the model results for organic compounds during both July and January 2013. Currently, the evaluation includes a comparison with OC observations only during January over only three locations of the relatively large model domain. Furthermore, please mention in the text what factor have you used to convert the modeled OA to OC. 3) The total PM2.5 measurements have been used to evaluate the model performance during July without explaining the rationale of this choice since the focus of this study is solely the organic fraction of the aerosols. I suggest removing the PM2.5 evaluation or at least moving it to the supplement. 4) Can you include more OC/OA observations over other areas of their domain in your evaluation? 5) It is also important to compare the simulated*

POA and SOA against observations (e.g., from AMS). Furthermore, it would be helpful to show how the model performance against SOA measurement changes between the BC and the S1, S2, S3 cases.

Response 13: (1) For the updates in CMAQ except for water partition into OPM and non-ideality of the organic-water mixture, previous studies have extensively examined the model performances and will not be further discussed in detail in this work. The text has been revised in L149-161 to reflect this:

"**Model configurations were largely based on that used by Hu et al. (2016) as summarized below. Firstly, SAPRC-11 was expanded for a more detailed treatment of isoprene oxidation and tracking dicarbonyl products (glyoxal and methylglyoxal) from different groups of major precursors (Ying et al., 2015). Secondly, SOA from isoprene epoxydiols (IEPOX), methacrylic acid epoxide (MAE) and dicarbonyls through surface-controlled irreversible reactive uptake were added (Hu et al., 2017; Li et al., 2015; Liu et al., 2020; Ying et al., 2015). Thirdly, the heterogeneous formation of secondary nitrate and sulfate from NO_2 and SO_2 reaction on the particle surfaces (Ying et al., 2014) were added, which is an important source of secondary inorganic aerosols (Zheng et al., 2015) and improves model estimates of nitrate and sulfate (Qiao et al., 2018; Shi et al., 2017). Fourthly, SOA yields were corrected for vapor wall loss (Zhang et al., 2014). Impacts of the above updates on model performances have been extensively discussed in the cited work and will not be further investigated in the current study.**"

(2) Unfortunately, detailed chemical composition measurements for aerosols are very limited in China during 2013. We only have observations of OC and OA in January of 2013 and $PM_{2.5}$ in July of 2013 available for model evaluation. Thus, we opt to use the most relevant data to provide a very limited assessment of the capability of the model in predicting SOA. The factors for OA to OC conversion follow the OM:OC ratio listed in Table S1 and S2 for SOA. POC is directly predicted by the model. The text has been revised in L291-292 to make this clear:

"**The factors used to convert SOA to OC (SOC) are listed in Table S1-S2. OC from POA (POC) is directly predicted by the model.**"

(3) Even though limited OC/OA measurements are available to us during this period, the base case model is later applied by another research group to model wintertime SOA formation in east China (Liu et al., 2020). The predicted OC and SOA agree well with observations (Figure 2 of Liu et al. 2020), and the model performance statistics for OC and SOA are similar to those of $PM_{2.5}$. We agree with the reviewer that $PM_{2.5}$ is not an ideal indicator to evaluate the capability of the model in predicting SOA, however, as a significant fraction of $PM_{2.5}$ in July is secondary, this still provides an indirect assessment of the model prediction of oxidation capacity of the atmosphere, which is import for

SOA formation. Additional modeling studies are needed to evaluate the performance of the model in summer.
We have explained this in the revised text (L327-329) as following:
“**Due to the lack of observed OC and OA in July of 2013, as an alternative, model performances are evaluated by comparing predicted and observed $PM_{2.5}$ at ground sites (Figure S1) as shown in Figure S3.**”

(4) We do not have more OC and OA data for the simulating episode of 2013.

(5) We did not have SOA observations in this episode. We compared the modeled SOA/POA ratio with AMS observations from other literature, finding a significant underestimation in the current model. This bias might be due to missing SOA from combustion (intermediate volatile organic compounds, IVOCs) and not treating POA as semi-volatile. We added a discussion about the model bias in the revised manuscript in L309-326:

“**Again, no apparent changes of SOA nor OA are observed between case S3 and BS (not shown), since POA is predicted to be the primary contributor to OA at Beijing in winter in the current model, with an averaged SOA/POA ratio of 0.12. This ratio is much lower than the field observation of about 0.45-1.94 (Zhao et al., 2019; Sun et al., 2013; Sun et al., 2016). The bias might be due to the missing SOA converted by partitioning and aging of semi-volatile POA as well as oxidation from intermediate volatile organic compounds (IVOCs) and VOC oxidation products. Those pathways are shown to be important for SOA formation by modeling, field and chamber studies (Hodzic et al., 2010; Jimenez et al., 2009; Murphy et al., 2017; Robinson et al., 2007; Shrivastava et al., 2008; Tkacik et al., 2012; Zhao et al., 2014; Zhao et al., 2016a).**

A sensitivity test was performed by using the newest CMAQ model version 5.3.1 that includes all the above processes in the aerosol module. The SOA/POA ratio in Beijing is improved greatly to be 1.53 in winter. However, high uncertainties still exist in the emissions of the involved precursors and characterization of SOA formation through these processes, needing further constrains by observations. Their influences on water partitioning into OPM and non-ideality of the organic-water mixture on SOA will be evaluated in a future study.”

Since SOA is underestimated, no significant differences in BS and S3 are observed. Case S1 and S2 are designed for the sensitivity test of model results to water partitioning and non-ideality of condensed organics separately. Therefore, we did not evaluate model performances from S1 and S2.

Comment 14: *Page 10 lines 251-253: The authors state here that the impacts of water-cocondensation and polarity of organic condensed species on SOA* *formation are not significant during winter. This highlights the need to evaluate their model results during July where they have found significant changes with the basecase simulation. Furthermore, the results from the three sensitivity simulations should be evaluated individually.*

Response 14: Unfortunately, we have no observations of OC, OA or (and) SOA of July 2013. We tried to evaluate model performances by comparing the predicted and observed $PM_{2.5}$ as an alternative. This has been explained in the revised manuscript in L327-329:

"**Due to the lack of observed OC and OA in July of 2013, as an alternative, model performances are evaluated by comparing predicted and observed PM2.5 at ground sites (Figure S1) as shown in Figure S3.**"

Since SOA is underestimated, no significant differences in BS and S3 are observed. Case S1 and S2 are designed for the sensitivity test of model results to water partitioning and non-ideality of condensed organics separately. Therefore, we did not evaluate model performances from S1 and S2.

Comment 15: *Page 10 lines 254-256: These are indeed possible factors. Can the authors comment, based on their analysis, which of these two possible factors is more important and try to be more specific? A comparison with AMS observations would be helpful here.*

Response 15: We did a sensitivity test by simulating the same episode with the current CMAQv5.3.1 in which POA was treated as semi-volatile and aging in the gas phase. Also, a missing source of SOA from intermediate VOCs (IVOCs) oxidation and aging of IVOCs and VOCs oxidation products (pcSOA) was added in CMAQv5.3.1. The results showed that the modeled SOA/POA has been improved from 0.12 of case S3 to 1.53 (Figure R1), more close to AMS observations. However, there were still some peak values underestimated by the model, which might be due to the uncertainties of POA emissions. We have revised the text (L309-326), as mentioned in Response 13 (5).

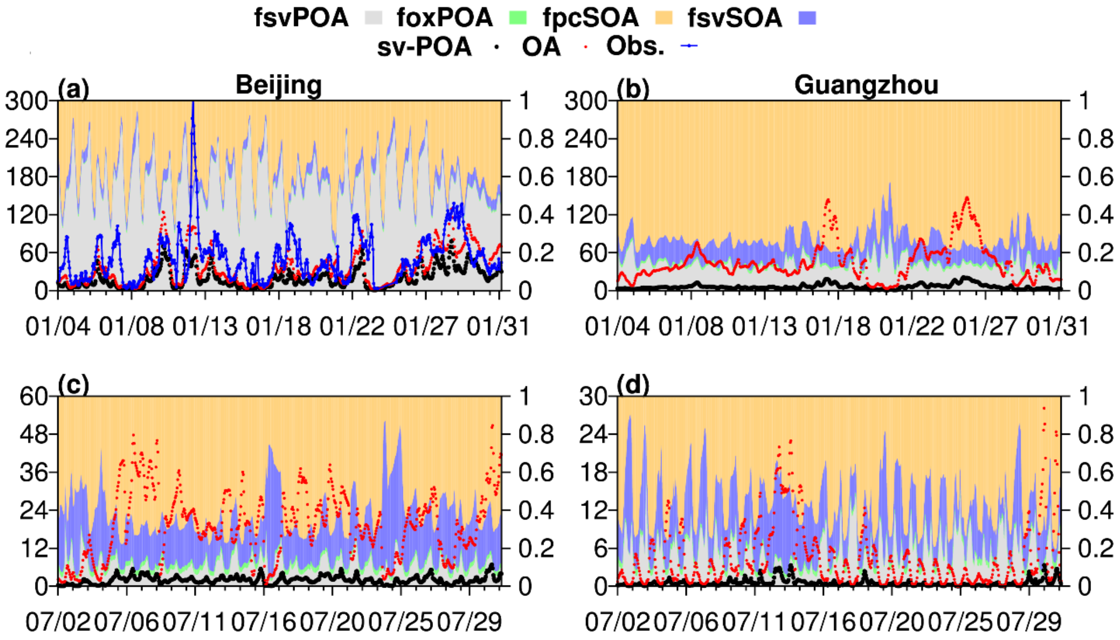

Figure R1. Modeled concentration of semi-volatile POA (sv-POA), OA and fraction of each organic aerosol component fsvPOA (sv-POA), foxPOA (oxidation of sv-POA), fpcSOA (pc-SOA) and fsvSOA (traditional SOA) in Beijing (a, c) and Guangzhou (b, d). Observations of OA in January 2013 at Beijing (Obs.) are also included in (a). The left axis is the concentration (μg m^{-3}) and the right axis is the fraction of OA components.

Comment 16: *Page 11 line 263: The aging of POA, under specific conditions can enhance the SOA formation, especially over polluted areas. Can the authors comment how this important omission of their model configuration can affect their result? Once again, a comparison against POA and SOA from AMS observations will be helpful to identify the limitations of their model due to the treatment of POA as non-volatile and non-reactive.*

Response 16: We did a sensitivity test by CMAQv5.3.1 that includes SOA from POA aging, IVOCs oxidation, and aging of IVOCs and VOCs oxidation products. The modeled SOA/POA is improved greatly in Beijing with no significant improvement in OA compared to the results of case S3. A significant improvement of SOA was observed from the contribution from IVOCs oxidation, and the aging of IVOCs and VOCs oxidation products (pc-SOA). Discussions about this have been included in the revised text in L309-326, as mentioned in Response 15.

Also, we examined the sensitivity of SOA and organic liquid water (ALW_{org}) to pcSOA and POA in an offline calculation in Beijing, Guangzhou, Jinan, and Nanjing. POA has the same properties as we used in the model. Non-volatile isoprene SOA is taken to represent pcSOA as their similarities in saturation vapor pressure and O:C ratio. We found that both SOA and ALW_{org} are positively correlated with pcSOA, increased by 2-5 times in different locations when pcSOA increased by 2 times.

Comment 17: *Page 11 line 264: Please provide two spatial maps of the fraction SOA/Total OA during January and July 2013 so as to show the contribution of POA and SOA to total OA during each simulation period.*

Response 17: We have added a figure of SOA/OA ratio by case BS and S3 (as shown below) in the revised supplemental materials (Figure S13).

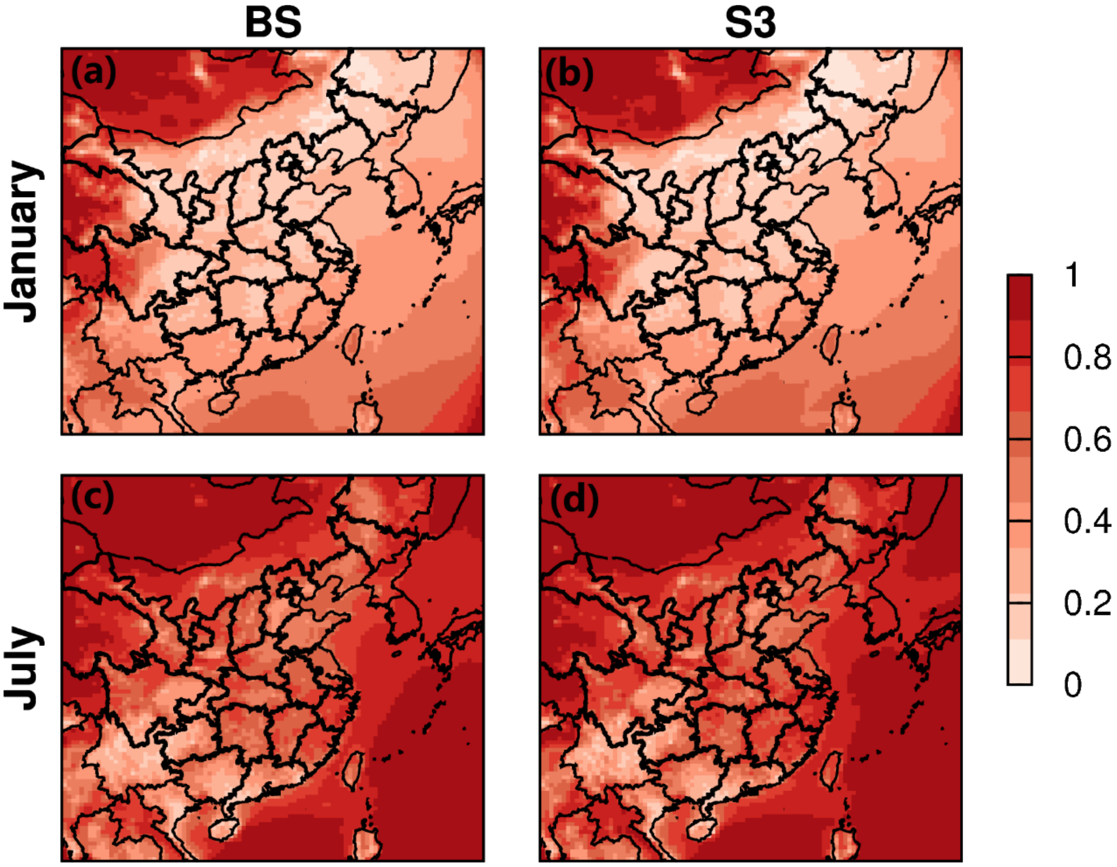

Figure R2. Averaged SOA/OA ratio from case BS and S3 during January and July of 2013.

Comment 18: *Page 13 line 317: You need a zonal map to show how the water partition changes with altitude and not the total column.*

Response 18: Most of the SOA and ALW_{org} retain in the lower levels of the troposphere. The information on altitude variation may not be very useful. Thus, no changes were made regarding this comment. We have also deleted this sentence in the revised manuscript to avoid confusion.

Comment 19: *Page 13, lines 322-336: It is not clear how you calculate the κ_{org} in your model. This is very important for this section.*

Response 19: We explained this in the revised text in L214-216:

“**Since ALW_{org} in this study is calculated mechanistically using the partitioning theory, κ_{org} can be estimated by rearranging Eq3:**

$$\kappa_{org} = \frac{ALW_{org}}{\rho_w V_{org}} \times \frac{1-a_w}{a_w} \tag{Eq4}$$

”

Comment 20: *Figure 1: The quality of the figure is poor. It is extremely difficult to see all the plotting data and the changes due to the use of different scenarios (especially in figure 1b).*

Response 20: We removed the results of BS since they are very similar to those of S3. Now the figure has been revised as follows:

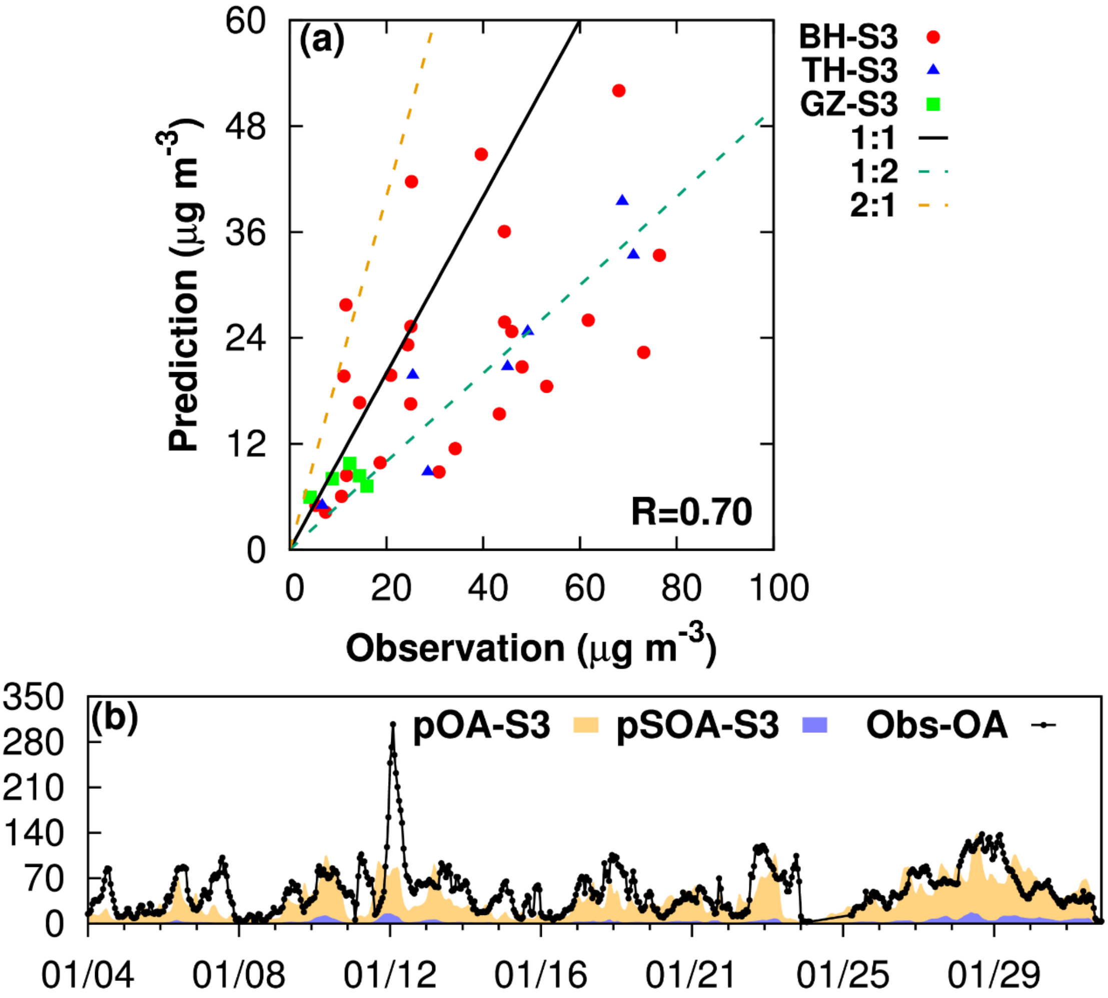

Figure 1. Comparison of (a) observed and modeled organic carbon concentration at University of Beihang (BH), Tsinghua University (TH) and Guangzhou (GZ); (b) observed organic aerosol (Obs-OA) at Beijing and predictions of total OA (pOA) and SOA (pSOA), unit is µg m-3. Locations of monitoring sites are shown in Figure S1.

Comment 21: *Figure 2: I found the use of daily maximum concentration in the* *"difference" maps misleading. Given that you have the monthly average SOA from the basecase simulation, I would prefer to see the absolute (and relative) change of the monthly average SOA due to the use of S3 as well, and not the daily maximum. Furthermore, please add (a), (b), etc. to each subplot of the figure and add this information to the figure caption (apply this change in the rest of the figures as well).*

Response 21: The monthly-averaged daily maximum differences have been replaced by the monthly-averaged differences for all the corresponding figures to reflect the general impacts on SOA and ALW_{org}. Each panel of the figure is labeled in sequence.

Comment 22: *Figure 4: All the fitted correlations listed here suggest that compounds with very low or zero O:C have negative hygroscopicity. Can you comment on this limitation and include a discussion in the text?*

Response 22: The relatively low values of hygroscopicity for low O:C ratio might be due to the linear regression. We also did an exponential fitting for the two variables so that the hygroscopicity falls in the range of (0,1) and is positively correlated with O:C ratio. The text and figure 4 have been revised accordingly.

In L402-407:

"**In both seasons, κ_{org} approaches zero and negative values as O:C decreases, which might be due to the linear regression of κ_{org} and O:C. To avoid this, an exponential fitting of the two variables is performed so that κ_{org} falls in the range of (0,1) and is positively correlated with O:C. In this case, the fitted correlations are $\kappa_{org}=1-\exp(-(O:C/1.88)^{2.29})$ and $\kappa_{org}=1-\exp(-(O:C/1.06)^{4.50})$ for January and July of 2013, respectively.**"

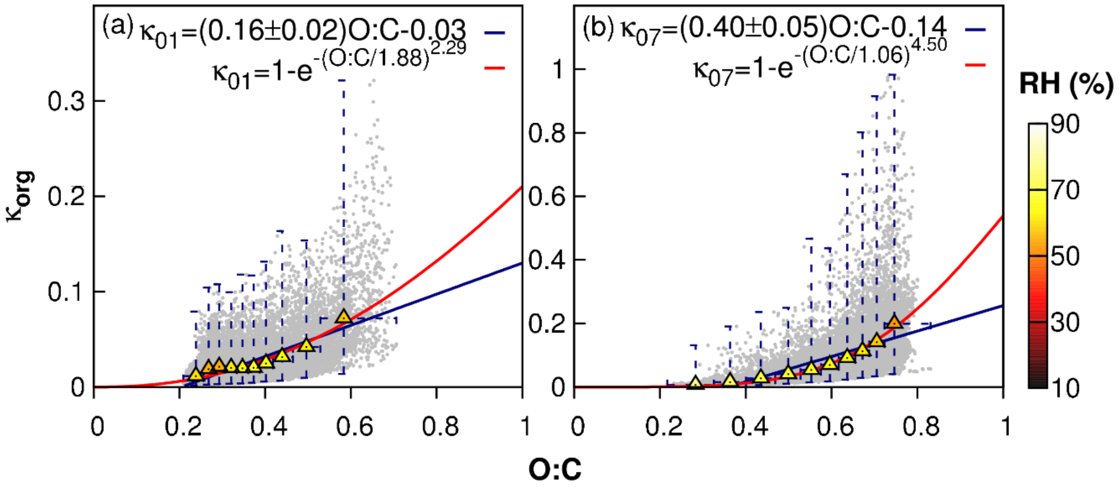

Figure 4. The correlation of hygroscopicity of organic aerosol (κ_{org}) and O:C ratio at 9 representative cities including Shenyang (SS), Beijing (BJ), Jinan (JN), Zhengzhou (ZZ), Xi'an (XA), Nanjing (NJ), Shanghai (SH), Chengdu (CD), and

Guangzhou (GZ) in January (a) and July (b) of 2013. O:C ratios are categorized into 10 bins. In each bin, the ranges of O:C and κ_{org} are represented by bars. The mean values of O:C and κ_{org} are represented by triangles colored by the averaged RH of each bin. The relationship between κ_{org} and O:C is fitted by a linear function with reduced major axis regression (blue lines) and an exponential function (red lines), respectively. κ_{01} and κ_{07} represent the fitted correlation for January and July, respectively.

Comment 23: *Table S3. Please explain in the table what the fraction in the last column stands for. How have you estimated the molecular weight and fraction of the POA from unknown compounds?*
Response 23: The last column is the molar fraction of each POA surrogate. We have clarified this in the first row of this column in the table. The molecular weight and molar fraction of unknown compounds of POA have been listed in the original table already, which are 390 and 0.3, respectively.

Minor Comments
Comment 24: *The language and structure of the sentences can be substantially improved in many parts of the manuscript. Just a few examples are listed here, but I suggest revising thoroughly the wording in the whole text.*
Response 24: We thank the reviewer for pointing this out. The whole text, as well as figures and tables, have been revised carefully.

Comment 25: *Page 5, lines109-11: Please rephrase. The sentence sounds wrong.*
Response 25: The text has been revised in L109-111 as follows:
"**Laboratory and field studies have observed water absorbed by SOA from a variety of precursor VOCs (Lambe et al., 2011; Zhao et al., 2016b; Asa-Awuku et al., 2010; Varutbangkul et al., 2006).**"

Comment 26: *Page 5, lines117-118: Please rephrase.*
Response 26: The sentence has been revised in L116-117 as follows:
"**The total water content is the summation of water associated with each solute at the same water activity.**"

Comment 27: *Page 6 line 144: OC and OA abbreviations have not been used before in the main text.*
Response 27: OA abbreviation has been explained in a previous part of the revised manuscript in L120-122:
"**Pye et al. (2017) found that the modeled organic aerosol (OA) improved significantly but biased high at nighttime when ALW_{org} is included in the calculation.**"

Thus, this sentence has been revised in L139-141 as follows:

“**The model performance was evaluated against observed meteorological parameters (temperature and relative humidity, RH) as well as $PM_{2.5}$, organic carbon (OC), and OA at ground monitoring sites.**”

Comment 28: *Page 8, line 204: Please correct the “can estimated” to “can be estimated”.*
Response 28: The text has been revised as instructed.

Comment 29: *Page 8 line 206: I would use the word “correlate” instead of “dependent”*
Response 29: The text has been revised in L219-229 as follows:

“**In many studies, κ_{org} is assumed to increase linearly with the oxidation state of OA, expressed as the O:C ratio (Massoli et al., 2010;Duplissy et al., 2011;Lambe et al., 2011).**”

Comment 30: *Page 9 line 229: Change “in” with “on”*
Response 30: The text has been revised as instructed.

Comment 31: *Page 10 line 254: Change the sentence to: “In Beijing and Guangzhou, these impacts are not significant during winter”*
Response 31: The sentence has been revised in L298-300 as:
“**No significant differences in OC are observed in S3 compared to BS (not shown), likely due to the biased-low SOA predicted in the current model so that limits the impact of ALW_{org} on SOA formation.**”

Comment 32: *Page 11 line 274-275: Please rephrase.*
Response 32: The sentence has been revised as follows:
“**The criteria of MFB and MFE followed recommendations by Boylan and Russell (2006).**”

Comment 33: *Page 11 line 282: Which two areas? You have mentioned several areas in the previous sentence.*
Response 33: The sentence has been revised as follows:
“**Monthly-averaged SOA concentrations in the above areas are up to 25 and 15-20 μg m^{-3}, respectively.**”

Comment 34: *Page 13 line 317: “column water”. Please rephrase*
Response 34: The text has been revised in L378 as follows:
“**Based on the column concentrations of ALW_{org} and ALW_{org}/SOA ratio (Figure S8),**”

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